AMENDMENT(S) TO THE CLAIMS

This listing of claims will replace all prior versions and listings of claims in this application:

Listing of Claims:

1. (Currently amended) A method for increasing the hardness of silica/rubber mixtures comprising blending with said mixture at least one silane and a hardness-increasing amount of at least one member selected from the group consisting of thixotropic fumed silica, or precipitated silica wherein the total amount of silica is above 100 phr , an MQ resin wherein Q is $SiO_{4/2}$, M is $R^aR^bR^cSiO_{1/2}$, and R^a, R^b, and R^c are the same or different functional or non-functional organic groups, ~~carbon black~~, a thermoplastic resin selected from the group consisting of high-density polyethylene, ultra high molecular weight polyethylene and low density-polyethylene and a thermosetting resin, wherein the silane is a blocked ~~or unblocked~~ mercaptosilane, and wherein the silica/rubber mixture optionally includes inorganic filler,

and wherein said hardness increasing amount is ~~from 1 to 100 phr~~ above the amount necessary to achieve equivalent Shore A hardness of the silica/rubber mixture as compared with the use of bis-(triethoxysilylpropyldisulfide) as the silane, and, ~~wherein the total amount of the member including any inorganic filler is above 100 phr and up to about 160 phr~~

wherein the blocked mercaptosilane has a formula selected from the group consisting of:

$$[[(ROC(=O))_p\text{-}(G)_i]_k\text{-}Y\text{-}S]_r\text{-}G\text{-}(SiX_3)_s \quad (1)$$

and

$$[(X_3Si)_q\text{-}G]_a\text{-}[Y\text{-}[S\text{-}G\text{-}SiX_3]_b]_c \quad (2),$$

wherein

Y is a polyvalent species $(D)_zA'(=E)$, each wherein the atom (A') attached to the unsaturated heteroatom (E) is attached to the sulfur, which in turn is linked via a group G to the silicon atom;

each R is chosen independently from hydrogen, straight, cyclic, or branched alkyl that may or may not contain unsaturation, alkenyl groups, aryl groups, and aralkyl groups wherein eacr R contains from 1 to 18 carbon atoms;

each G is independently a monovalent or polyvalent group derived by substitution of alkyl, alkenyl, aryl, or aralkyl wherein G can contain from 1 to 18 carbon atoms, with the proviso that G is not such that the silane would contain an α,β-unsaturated carbonyl including a carbon-carbon double bond next to the thiocarbonyl group, and if G is monovalent wherein p = 0, G can be a hydrogen atom;

X is independently a group selected from the group consisting of -Cl, -Br, RO-, $RC(=O)O$-, $R_2C=NO$-, R_2NO- or R_2N-, -R, $-(OSiR_2)_l(OSiR_3)$ wherein each R is as above and at least one X is not -R;

D is oxygen, sulfur, or (-NR-);

A' is carbon, sulfur, phosphorus, or sulfonyl;

E is oxygen, sulfur, or NR;

p is 0 to 5; r is 1 to 3; z is 0 to 2; q is 0 to 6; a is 0 to 7; b is 1 to 3; j is 0 to 1, but it may be 0 only if p is 1; c is 1 to 6; t is 0 to 5; s is 1 to 3; k is 1 to 2, with the provisos that

(A) if A' is carbon, sulfur, or sulfonyl, then

(i) a + b = 2 and

(ii) k = 1;

(B) if A' is phosphorus, then a + b = 3 unless both (i) c > 1 and (ii) b = 1, in which case a = c + 1; and

(C) if A' is phosphorus, then k is 2 .

2. (Canceled)

3. (Canceled)

4. (Currently amended) The method of claim ~~2~~ 1 wherein the rubber is selected from the group consisting of solution styrene-butadiene rubber, emulsion styrene-butadiene rubber, natural rubber, polybutadiene, ethylene-propylene co- and terpolymers, acrylonitrile-butadiene rubber, isoprene, polystyrene and poly α-methyl styrene, cis-1,4-polyisoprene rubber, styrene/butadiene copolymer rubber, 3,4-polyisoprene rubber, isoprene/butadiene rubber, styrene/isoprene/butadiene terpolymer rubber, cis-1,4-polybutadiene, vinyl polybutadiene rubber, styrene/isoprene copolymers, emulsion polymerization prepared styrene/butadiene/acrylonitrile

terpolymer rubber and butadiene/acrylonitrile copolymer rubber, emulsion polymerization prepared styrene/butadiene/acrylonitrile terpolymer rubbers containing 2 to 40 weight percent bound acrylonitrile in the terpolymer and combinations thereof.

Claim 5, (Cancelled).

6. (Original) The method of claim 1 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

7. (Original) The method of claim 1 wherein the silica/rubber mixture further comprises an inorganic filler.

8. (Previously presented) The method of claim 7 wherein the inorganic filler is selected from the group consisting of titanium dioxide, aluminosilicate, alumina, calcium carbonate, carbon fibers, glass fibers, kaolin clay, mica, talc and wollastonite.

9. (Currently amended) The method of claim 1 wherein the at least one member is thixotropic-(hydrophilic and hydrophobic) fumed (pyrogenic) silica.

Claims 10 to 13, (Cancelled).

14. (Original) The method of claim 9 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

15. (Original) The method of claim 9 wherein the silica/rubber mixture further comprises an inorganic filler.

16. (Previously presented) The method of claim 15 wherein the inorganic filler is selected from the group consisting of titanium dioxide, aluminosilicate, alumina, calcium carbonate, carbon fibers, glass fibers, kaolin clay, mica, talc and wollastonite.

17. (Original) The method of claim 1 wherein the member is precipitated silica.

Claims 18 to 21, (Cancelled).

22. (Original) The method of claim 17 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

23. (Original) The method of claim 17 wherein the silica/rubber mixture further comprises an inorganic filler.

24. (Previously presented) The method of claim 23 wherein the inorganic filler is selected from the group consisting of titanium dioxide, aluminosilicate, alumina, calcium carbonate, carbon fibers, glass fibers, kaolin clay, mica, talc and wollastonite.

25. (Currently amended) ~~The method of claim 1~~ A method for increasing the hardness of silica/rubber mixtures comprising blending with said mixture at least one silane and a hardness-increasing amount of an ~~wherein the member is the~~ MQ resin wherein Q is $SiO_{4/2}$, M is $R^aR^bR^cSiO_{1/2}$, and R^a, R^b, and R^c are the same or different functional or non-functional organic groups, wherein the silane is a blocked mercaptosilane, and wherein the silica/rubber mixture optionally includes inorganic filler,

and wherein said hardness increasing amount is above the amount necessary to achieve equivalent Shore A hardness of the silica/rubber mixture as compared with the use of bis-(triethoxysilylpropyldisulfide) as the silane.

Claims 26 to 29, (Cancelled).

30. (Original) The method of claim 25 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

31. (Original) The method of claim 25 wherein the silica/rubber mixture further comprises an inorganic filler.

32. (Previously presented) The method of claim 31 wherein the inorganic filler is selected from the group consisting of titanium dioxide, aluminosilicate, alumina, calcium carbonate, carbon fibers, glass fibers, kaolin clay, mica, talc and wollastonite.

33. (Currently amended) The method of claim 1 wherein the at least one member ~~is~~ further includes carbon black.

34. (Original) The method of claim 33 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

35. (Original) The method of claim 1 wherein the at least one member is ~~a~~ the thermoplastic resin.

36. (Original) The method of claim 35 wherein the thermoplastic resin is selected from the group consisting of high-density polyethylene, ultra high molecular weight polyethylene, and low density-polyethylene.

37. (Original) The method of claim 35 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

38. (Currently amended) The method of claim 1 wherein the at least one member is a thermosetting resin.

39. (Currently amended) The method of claim 1 wherein the ~~resin~~ at least one member is a high glass transition thermoplastic resin.

40. (Original) The method of claim 39 wherein the high glass transition resin is selected from the group consisting of polyphenylene sulfide, polyamide, polyimide, polyamide-imide, polycarbonate, nylons, and polymethylmethacrylate.

41. (Original) The method of claim 39 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

42. (Currently amended) An article of manufacture comprising a silica/rubber mixture hardened by blending with said mixture at least one silane and a hardness-increasing amount of at least one member selected from the group consisting of thixotropic fumed silica, or precipitated silica wherein the total amount of silica is above 100 phr , an MQ resin wherein Q is

$SiO_{4/2}$, M is $R^aR^bR^cSiO_{1/2}$, and R^a, R^b, and R^c are the same or different functional or non-functional organic groups, ~~carbon black~~, a thermoplastic resin selected from the group consisting of high-density polyethylene, ultra high molecular weight polyethylene and low density-polyethylene and a thermosetting resin, wherein the silane is a blocked ~~or unblocked~~ mercaptosilane, and wherein the silica/rubber mixture optionally includes inorganic filler,

and wherein said hardness increasing amount is ~~from 1 to 100 phr~~ above the amount necessary to achieve equivalent Shore A hardness of the silica/rubber mixture as compared with the use of bis-(triethoxysilylpropyldisulfide) as the silane, and, ~~wherein the total amount of the member including any inorganic filler is above 100 phr and up to about 160 phr~~

wherein the blocked mercaptosilane has a formula selected from the group consisting of:

$$[[(ROC(=O))_p\text{-}(G)_i]_k\text{-}Y\text{-}S]_r\text{-}G\text{-}(SiX_3)_s \quad (1)$$

and

$$[(X_3Si)_q\text{-}G]_a\text{-}[Y\text{-}[S\text{-}G\text{-}SiX_3]_b]_c \quad (2),$$

wherein

Y is a polyvalent species $(D)_zA'(=E)$, each wherein the atom (A') attached to the unsaturated heteroatom (E) is attached to the sulfur, which in turn is linked via a group G to the silicon atom;

each R is chosen independently from hydrogen, straight, cyclic, or branched alkyl that may or may not contain unsaturation, alkenyl groups, aryl groups, and aralkyl groups wherein eacr R contains from 1 to 18 carbon atoms;

each G is independently a monovalent or polyvalent group derived by substitution of alkyl, alkenyl, aryl, or aralkyl wherein G can contain from 1 to 18 carbon atoms, with the proviso that G is not such that the silane would contain an α,β-unsaturated carbonyl including a carbon-carbon double bond next to the thiocarbonyl group, and if G is monovalent wherein p = 0, G can be a hydrogen atom;

X is independently a group selected from the group consisting of -Cl, -Br, RO-, RC(=O)O-, R_2C=NO-, R_2NO- or R_2N-, -R, -$(OSiR_2)_l(OSiR_3)$ wherein each R is as above and at least one X is not -R;

D is oxygen, sulfur, or (-NR-);

A' is carbon, sulfur, phosphorus, or sulfonyl;

E is oxygen, sulfur, or NR;

p is 0 to 5; r is 1 to 3; z is 0 to 2; q is 0 to 6; a is 0 to 7; b is 1 to 3; j is 0 to 1, but it may be 0 only if p is 1; c is 1 to 6; t is 0 to 5; s is 1 to 3; k is 1 to 2, with the provisos that

(A) if A' is carbon, sulfur, or sulfonyl, then

(i) a + b = 2 and

(ii) k = 1;

(B) if A' is phosphorus, then a + b = 3 unless both (i) c > 1 and (ii) b = 1, in which case a = c + 1; and

(C) if A' is phosphorus, then k is 2 .

43. (Original) The article of claim 42 wherein the silane is 3-octanoylthio-1-propyltriethoxysilane.

44. (Currently amended) ~~The article of claim 43~~ An article of manufacture comprising a silica/rubber mixture hardened by blending with said mixture at least one silane and a hardness-increasing amount of an ~~wherein said member is the~~ MQ resin wherein Q is $SiO_{4/2}$, M is $R^aR^bR^cSiO_{1/2}$, and R^a, R^b, and R^c are the same or different functional or non-functional organic groups, wherein the silane is 3-octanoylthio-1-propyltriethoxysilane, and wherein the silica/rubber mixture optionally includes inorganic filler, and wherein said hardness increasing amount is above the amount necessary to achieve equivalent Shore A hardness of the silica/rubber mixture as compared with the use of bis-(triethoxysilylpropyldisulfide) as the silane.

45. (Currently amended) The article of claim 44 wherein said article is a tread portion of a tire.